Metal-to-insulator crossover and pseudogap in single-layer compound $Bi_{2+x}Sr_{2-x}Cu_{1+y}O_{6+\delta}$ single crystals in high magnetic fields

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The in-plane $\rho_{ab}(H)$ and the out-of-plane $\rho_c(H)$ magneto-transport in magnetic fields up to 28 T has been investigated in a series of high quality, single crystal, hole-doped La-free Bi2201 cuprates for a wide doping range and over a wide range of temperatures down to 40 mK. With decreasing hole concentration going from the overdoped (p=0.2) to the underdoped (p=0.12) regimes, a crossover from a metallic to and insulating behavior of $\rho_{ab}(T)$ is observed in the low temperature normal state, resulting in a disorder induced metal insulator transition. In the zero temperature limit, the normal state ratio $\rho_c(H)/\rho_{ab}(H)$ of the heavily underdoped samples in pure Bi2201 shows an anisotropic 3D behavior, in striking contrast with that observed in La-doped Bi2201 and LSCO systems. Our data strongly support that that the negative out-of-plane magnetoresistance is largely governed by interlayer conduction of quasiparticles in the superconducting state, accompanied by a small contribution of normal state transport associated with the field dependent pseudogap. Both in the optimal and overdoped regimes, the semiconducting behavior of $\rho_c(H)$ persists even for magnetic fields above the pseudogap closing field H_{pg}. The method suggested by Shibauchi *et al.* (Phys. Rev. Lett. **86**, 5763, (2001)) for evaluating H_{pg} is unsuccessful for both under- and overdoped Bi2201 samples. Our findings suggest that the normal state pseudogap is not always a precursor of superconductivity.

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I. INTRODUCTION

The electrical transport, notably the interlayer transport, of the layered high-T_c superconductors (HTS) shows anomalous properties related to the quasi two-dimensional structure which have been studied very extensively in recent years. In the normal state the interlayer conductivity gives information on the quasiparticle properties[1]. This behavior of the quasiparticles is anomalous in the normal state of the HTS, which is of importance for elucidating the yet to be understood mechanism of superconductivity. The understanding of the fundamental interlayer transport properties of HTS is a challenging physical problem in its own right.

One of the unusual features of the normal-state properties is the coexistence of a metallic-like temperature dependence of the in-plane resistivity ρ_{ab} and a semiconducting-like behavior for the out-of-plane resistivity ρ_c (see e.g. Refs. [2,3,4]). The very different behavior of the resistivities ρ_{ab} and ρ_c implies a 2D confinement and is *a priori* incompatible with a Fermi-liquid behavior[5]. Over the last few years, many theoretical and experimental investigations have been devoted to the transport properties of HTS. In particular, in the temperature region showing the semiconducting-like c-axis resistivity, most compounds reveal a negative out-of-plane magnetoresistance: the $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212)[1,6,7,8,9,10], the $La_{2-x}Sr_xCuO_4$ (LSCO)[11,12], and the La-doped $Bi_2Sr_{2-x}La_xCuO_{6+\delta}$ (BSLCO) system[5,13].

The observed semiconducting-like $\rho_c(T)$ and negative out-of-plane magnetoresistance have been discussed in terms of different models, such as c-axis tunneling with a strong suppression by charge fluctuations excited in the process of tunneling[14], c-axis hopping with interplanar scattering[12], a reduction of the density of states due to superconducting fluctuations[6,8,9], and a pseudogap and/or spin gap opening in the density of states[7,11]. This is another striking feature of HTS. Of particular interest in the physics of carriers in strongly correlated and disordered systems to which HTS belong is the coexistence of superconductivity and localization. The latter phenomenon is one further peculiarity of HTS. Disorder in a metallic system can cause localization of the electronic states and lead to a metal-insulator transition[15].

The metal-insulator transition has been observed in the superconducting systems LSCO[16] and $Pr_{2-x}Ce_xCuO_{4+\delta}$[17] at optimal doping and BSLCO well inside the underdoped regime[18]. The insulating behavior in these systems is characterized by an in-plane resistivity $\rho_{ab}(T)$ which increases as $\log(1/T)$. These results demonstrate that the metal-insulator crossover in cuprates should not be universally associated with doping but rather with the observation of a unified $\log(1/T)$ temperature dependence of the resistivity suggesting a peculiar charge localization in the above mentioned cuprates[18].

It is difficult to obtain an overall picture of the metal-insulator transition in cuprates because only three systems have been studied, with strikingly different results obtained for the metal-insulator crossover for BSLCO when compared to those for LSCO and $Pr_{2-x}Ce_xCuO_{4+\delta}$. The anomalous transport should be more noticeable in the vicinity of the metal-insulator

transition and in the $T \to 0$ limit, suggesting the existence of a close link between charge transport and strong electron correlation. However, up to now the behavior of cuprates in the normal state in the $T \to 0$ limit remains an open issue.

One of the unresolved, but all-important issues of high temperature superconductivity, is the connection of normal state correlations cited above, and referred to as a pseudogap, to the origins of the high T_c[19]. Many experiments (e.g. nuclear magnetic resonance[20], photoemission[19], tunneling[21]) have provided evidence that in the normal state of underdoped HTS, a pseudogap exists in the electronic excitation spectra below a temperature $T^* > T_c$. This leads to a semiconducting-like behavior of the c-axis resistivity below T^*. Photoemission experiments (ARPES) have seen d-wave symmetry in the pseudogap structure[19]. In scanning tunneling measurements on Bi2212, Renner *et al.*[21] have found this pseudogap to be present both in underdoped and overdoped samples, and to scale with the superconducting gap. Certain groups have proposed, that the pseudogap in the normal state can be seen as a precursor for the occurrence of superconductivity where the superconducting phase-coherence is suppressed by thermal or quantum fluctuations, e.g. Refs.[22,23,24]. More recently, from interlayer tunneling spectroscopy in the Bi2212 system, evidence for a definite difference between the superconducting gap and the pseudogap has been obtained[25]. This result is further reinforced by nuclear magnetic resonance measurements[26] on the underdoped cuprate $YBa_2Cu_4O_8$ ($T_c = 74$ K) which showed that a magnetic field of 23 T, while reducing T_c by 23%, has no effect on the pseudogap, suggesting that it has a distinct origin from that of the superconductivity.

In the case of a non-superconducting origin, a pseudogap can be formed in the spin-part of the excitation spectrum in the context of spin charge separation. In studies of the magnetic field dependence of the spin gap in the near optimally doped $YBa_2Cu_3O_7$ in the normal state[27,28] probed using the spin lattice relaxation rate, contradictory results were obtained. On the one hand, in an intensive study of the anisotropic transport on the Bi2212 system[29] the authors found that the onset of semiconducting-like $\rho_c(T)$ does not coincide with the opening of the spin gap seen in the in-plane resistivity $\rho_{ab}(T)$. The pseudogap opening temperature, on the other hand, coincides with the onset of the semiconducting-like behavior observed in $\rho_c(T)$ on the $YBa_2Cu_3O_7$ system. Since the normal-state properties in the high-T_c superconductors are known to depend strongly on the carrier concentration, the reported transport and magnetotransport data in the normal state cannot be easily categorized to form a common picture. There is currently no consensus concerning at what temperature the pseudogap opens[30]. An experimental investigation of the possible correlation between the pseudogap and the out-of-plane magnetoresistance in layered HTS at high magnetic fields is therefore of crucial importance.

In previous measurements[31] we have studied the c-axis magnetoresistance in the La-free $Bi_{2+x}Sr_{2-x}Cu_{1+y}O_{6+\delta}$ (Bi2201) single crystals with $T_c = 9.5$ K under magnetic fields up to 28 T and over a temperature range $6 - 100$ K. The observed isotropic behavior of the normal-state magnetoresistance with respect to the orientation of the magnetic field (perpendicular and parallel to the CuO_2 planes) shows that only the effect of the magnetic field on the spins (Zeeman effect) is important in the normal state. Such a result makes it difficult to explain the negative magnetoresistance with models based on superconductivity involving superconducting fluctuations or a pseudogap as a precursor of complete superconductivity. Shibauchi *et al.*[32] have reported c-axis resistivity measurements in fields up to 60 T in underdoped and overdoped Bi2212 crystals, from which they made a first evaluation of the pseudogap closing field H_{pg}. These results again indicate the predominant role of spins over orbital effects in the formation of the pseudogap. However, because of the high $T_c = 67 - 78$ K and very high upper critical field, H_{c2}, for Bi2212 crystals the available 60 T field was insufficient to suppress superconductivity at low temperatures and to evaluate H_{pg}, the authors[32] were forced to extrapolate their data. Direct measurements of H_{pg} were performed only at $T > 95$ K. So far as little is known about the effect of magnetic field, the H dependence of the pseudogap in HTS remains highly controversial.

In this paper we present, to our knowledge, the first measured temperature dependence for both the in-plane ρ_{ab} and the out-of-plane ρ_c resistivities and magnetoresistivities $\rho_{ab}(H)$ and $\rho_c(H)$ in hole-doped La-free Bi2201 cuprate at under, and optimal doping concentrations, and over a wide range of temperature down to 40 mK. Due to the lack of a sufficient amount of Bi2201 single crystals and especially crystals with different doping levels, the transport properties of this system have not previously been investigated in detail. Owing to the low critical temperature of Bi2201, 25 T magnetic fields are sufficient to suppress superconductivity in these samples in the $T \to 0$ limit, even at optimal doping[33]. We have suppressed superconductivity in single crystals using a 28 T resistive magnet at the Grenoble High Magnetic Field Laboratory, in order to measure the in-plane R_{ab} and the out-of-plane R_c resistances in the normal state in magnetic fields applied perpendicular and parallel to the ab-plane.

II. EXPERIMENT

It is known, that the stoichiometric composition Bi2201 is an insulating phase, and that single-phase superconducting crystals can be obtained by replacing Sr with either Bi or La[34]. In a compound, the optimal cation states for Sr, La and Bi, are Sr^{2+}, La^{3+} and Bi^{3+}, respectively. Therefore, the substitution of triva-

TABLE I: Summary of the properties of the investigated single crystals determined as described in the text: The carrier concentration per Cu atom (p), actual cationic compositions (Bi:Sr:Cu), ratios Bi/Sr, critical temperature (T_c), lattice parameter (c), disorder parameter ($k_F l$), pseudo gap closing field (H_{pg}), and the functional form of the magnetic field dependence of $\rho_c(H)$.

p	Bi:Sr:Cu	Bi/Sr	T_c (K)	c ($\AA$)	$k_F l$	H_{pg} (T)	Functional form of $\rho_c(H)$
0.12	2.66:1.33:0.85	2.0	2.3	24.57	0.6	≥ 30	$\rho_c(H) \simeq \rho_{c0} + a_1 H$
0.13	2.62:1.38:0.87	1.9	3	24.575	7	≥ 30	$\rho_c(H) = \rho_{c0} + a_2 H + b_2 H^2$
0.16	2.39:1.61:1.02	1.48	9	24.59	20	$\simeq 21$	$\rho_c(H,T) = \rho_{c0} + a_3 \exp(-H/b_3 T)$
0.17[a]	2.31:1.69:1.12	1.37	9.6	24.61	-	-	-
0.2	2.10:1.90:1.14	1.1	6.7	24.63	49	$\simeq 16$	$\rho_c(H,T) = \rho_{c0} + a_4 \exp(-H/b_4 T)$

[a]Complete $\rho_{ab}(H)$ and $\rho_c(H)$ data is unavailable for this sample so that we are unable to estimate all parameters.

lent La or Bi for divalent Sr in the BSLCO or in the La-free Bi2201 samples reduces the hole concentration in the CuO_2 planes. For the Bi2201 samples, Fleming *et al.*[35] and Harris *et al.*[36] found that as the Bi/Sr ratio increases, and one moves toward the bottom of the phase diagram of the solid solution, the number of holes doped into the system decreases, which thus pushes the system towards the hole-underdoped regime. The lower T_c, together with the larger residual resistivity of Bi2201 in comparison with BSLCO (the maximum T_c is 38 K[18]) apparently suggests that the disorder due to (Sr,Bi) substitution is stronger in Bi2201 than the disorder due to (Sr,La) substitution[37].

We were able to make high quality single-phase superconducting $Bi_{2+x}Sr_{2-x}$ $Cu_{1+y}O_{6+\delta}$ single crystals in the range of $0.1 < x < 0.7$, provided that the Cu content was slightly increased[38,39]. The investigated Bi2201 single crystals were grown by a KCl-solution-melt free growth method. A temperature gradient along the crucible results in the formation of a large closed cavity inside the solution-melt. In this case, the crystals are not in direct contact with the solidified melt in the crucible, thereby avoiding thermal stresses during cool down. The crystals were grown in the temperature range $830 - 850$ °C. The crystals had a platelet-like shape and mirror-like surfaces. The several tens of crystals grown in such a cavity, when characterized, are found to have almost identical properties. The quality of the crystals was systematically verified by measurements of the dc resistance, ac susceptibility, X-ray diffraction and scanning electron microscopy. To summarize the properties of the investigated crystals, we have regrouped in Table I the data of p (carrier concentration per Cu atom), actual cationic compositions, ratios Bi/Sr, T_c, and lattice parameters c.

The X-ray diffraction measurements were performed using a double-axis diffractometer. A CuK_α radiation monochromatized by a pyrolytic graphite crystal was employed. Both θ- and 2θ-scans of the ($00l0$) sublattice reflections and the ($00l \pm 1$) satellite reflections were used to assess structural perfection. These measurements were carried out before and after low-temperature experiments in magnetic fields. The half-width of the sublattice reflections in the X-ray rocking curves for the optimally doped single crystals consisting of two or three blocks did not exceed 0.3°, whereas for the crystals consisting of one block only (with dimensions of only 0.3×0.3 mm^2) it was less than 0.1°. This value is close to a resolution limit of a diffractometer. Both the $(\theta - 2\theta)$- and θ- X-ray diffraction profiles of the sublattice show no detectable structural defects. Thus, it can be concluded that even the sublattice contains no small-angle boundaries. For example, the half-width of both the main profile (0016) and the satellite reflections (00151), (00151)' in the X-ray rocking curves for the heavily underdoped single crystal with p = 0.13 (with large the Bi excess) was about 0.2°.

The composition of the crystals was studied using a Philips CM-30 electron microscopy with a Link analytical AN-95S energy dispersion X-ray spectrometer. The actual cationic compositions of each investigated crystal were measured at several different places on the crystal and the scatter in the data was less than 7%. Complementary measurements of our Bi2201 single crystal composition performed at the Material Science Center, University of Groningen (The Netherlands) have shown that our crystals are slightly underdoped due to oxygen depletion.

The dimensions of the crystals were $(0.4 - 0.8)$ $mm \times$ $(0.5 - 1)$ $mm \times (3 - 10)$ μm. The T_c value of the crystals formed by our free growth method can be as high as 13 K. However, we have found that the highest quality superconducting Bi2201 single crystals have a very narrow range of values of the lattice parameters $a = 5.360 - 5.385$ $\AA$ and $c = 24.57 - 24.63$ $\AA$. In this case the T_c (midpoint) values of the crystals lie in the region $3.5 - 9.5$ K in agreement with previous studies[36,40]. The transition width defined by the 10% and 90% points of the superconducting transition of crystals ranged from 0.5 to 1.7 K.

It is known, that overdoping or underdoping of $Bi_2Sr_2CaCu_2O_{8+\delta}$ can be achieved by cation substitutions or by changes in the oxygen content[41,42]. However, in the low-T_c Bi-based phase Bi2201, we have found that it is difficult to change the number of holes, because it is difficult to change the oxygen content. We have performed many attempts to change the T_c of single crystals, after changing the doping level, by means of an annealing in oxygen or argon at different temperatures. However, a careful characterization of the annealed samples revealed

that changes in T_c greater than ± 1 K, were always accompanied by a severe degradation of the sample quality and the occurrence of phase inhomogeneity in agreement with previous studies[40]. Most likely this is due to the fact that our crystals are close to the decomposition line. For this reason, in the following measurements, we used only high quality *as-grown* single crystals. For the investigation, samples with different T_c values were obtained by growing crystals with a different Bi content.

A four-probe contact configuration, with symmetrical positions of the low-resistance contacts ($< 1\Omega$) on both ab-surfaces of the sample was used for the measurements of R_{ab} and R_c resistances. The temperature and magnetic field dependence of the resistances $R_{ab}(T, H)$ and $R_c(T, H)$ were measured using a lock-in amplifier driven at $\approx$10.7 Hz. The measured resistances were then transformed to the respective resistivities ρ_{ab} and ρ_c using the crystal dimensions and the ratio of R_2/R_1 in the thin sample limit of the Montgomery technique[43]. For the low temperature magnetotransport measurements, the crystals were placed directly inside the mixing chamber of a Kelvinox top-loading dilution fridge and studied with the magnetic field H applied either parallel or perpendicular to the c -axis. A configuration with $\mathbf{H} \perp \mathbf{J}$ and $\mathbf{H} \parallel \mathbf{J}$ for the in-plane transport current $\mathbf{J}$ was used. For the out-of-plane transport current, the magnetic field H was applied both parallel to the c-axis and parallel to the ab-plane in the longitudinal ($\mathbf{H} \parallel \mathbf{c} \parallel \mathbf{J}$) and transverse ($\mathbf{H} \perp \mathbf{c} \parallel \mathbf{J}$) configurations.

The carrier concentration per Cu atom, p, in the Bi-based HTS cannot be unambiguously determined because the Bi ion does not have a fixed valency[44]. However, Ando *et al.*[45] have shown that the normalized Hall coefficient $R_H eN/V_0$ of various cuprates agree well in the temperature range 150 - 300 K and the data of $La_{2-x}Sr_xCuO_4$, for which p is unambiguous, can be used to estimate the doping level in other systems. Here e, N and V_0 are the electronic charge, the number of Cu atoms in the unit cell and the volume associated with each Cu atom, respectively.

In order to estimate the carrier concentration in our samples, following the method proposed by Ando *et al.*[45], we have measured the Hall coefficient R_H in several crystals[46,47] and compared the magnitudes of the normalized Hall coefficient[47] with the values reported for LSCO[48]. Subsequently, we estimated p in other samples using the empirical (nearly linear) relation between the excess Bi, x, and p. In the inset of Fig. 1, we show the values of T_c (closed circles) plotted vs p for our Bi2201 single crystals (the dashed line is shown a guide to the eye). It was found that optimum doping occurs at $p \simeq 0.17$ below which $T_c(p)$ shows a rapid drop as for the BSLCO system[45]. As can be seen in Fig. 1, our samples are basically in the optimally doped and underdoped side of the phase diagram and the data show the well-known parabolic behavior.

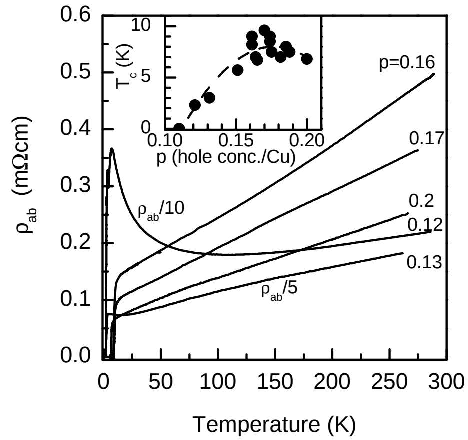

FIG. 1: Temperature dependence of the in-plane resistivity (ρ_{ab}) as function of temperature for Bi2201 samples with different hole concentrations. The inset shows the critical temperature for superconductivity (T_c) determined from ρ_{ab} as a function of hole concentration.

III. METAL-INSULATOR CROSSOVER AND ABSENCE OF A log(1/T) DIVERGENCE IN BOTH ρ_{ab} AND ρ_c

A. In-plane resistivity ρ_{ab}

In Fig. 1 (main panel) we show the temperature dependence of the in-plane resistivity ρ_{ab} for five single crystals with $T_c = 2.3$, 3, 6.7, 9.6 and 9 K (midpoint) at zero magnetic field for p values between 0.12 and 0.2. One can see that as for other cuprates, the magnitude of $\rho_{ab}(T)$ increases with decreasing carrier concentration. The resistivity curves give an almost linear temperature dependence for the optimally doped sample, positive curvature for the overdoped sample typical for other overdoped cuprates, and linear temperature dependence for the underdoped samples with a characteristic upturn at low temperatures ("semiconducting behavior").

Fig. 2 shows a semi-logarithmic plot of $\rho_{ab}(T)$ at various fixed magnetic fields for selected samples from Fig. 1 in order to emphasize the low-temperature behavior. Because the 20 and 27.5 T data are almost identical, we believe that we are measuring the true normal-state resistivity at our highest magnetic fields. ρ_{ab} for two underdoped samples, $p = 0.12$ (a) and 0.13 (b), goes through a minimum and then at temperatures $T \approx 30$ K (a) and $T \approx 10$ K (b), increases as $\log(1/T)$ as the temperature decreases, consistent with the onset of localization[49]. This behavior is in agreement with the results of Ono *et al.*[18], who found a logarithmic divergence of $\rho_{ab}(T)$ in underdoped BSLCO and LSCO samples. The $\log(1/T)$ dependence of $\rho_{ab}(T)$ reported by Ono *et al.*[18] extended over temperatures from 30 to 0.3 K without any sign of saturation at low temperatures. However, as can be seen from Fig. 2(a) and (b), ρ_{ab} in Bi2201 shows a downward deviation from a $\log(1/T)$ dependence at ultra low tem-

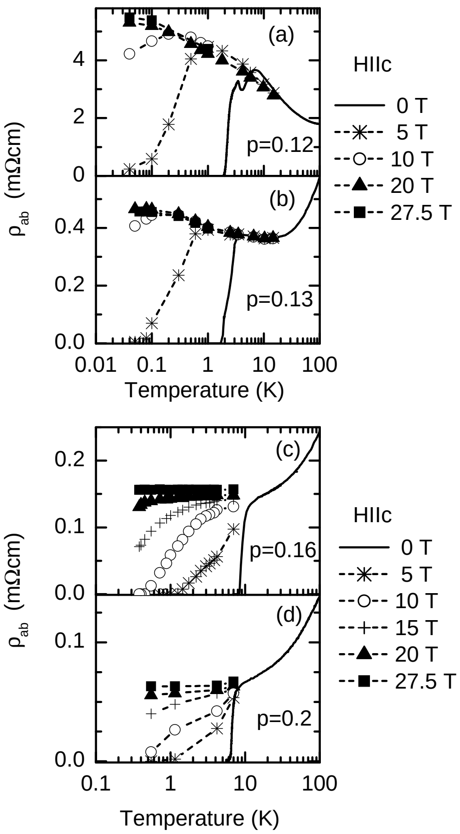

FIG. 2: Semi-log plot of ρ_{ab} versus temperature for various magnetic fields applied along the c-axis for four Bi2201 samples with different hole concentrations

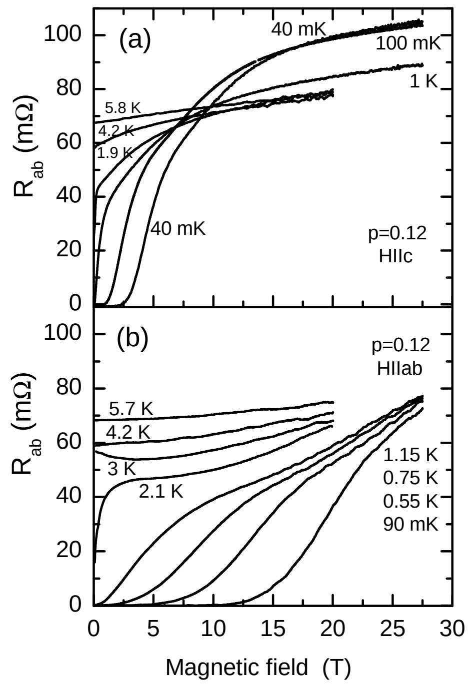

FIG. 3: In-plane resistance as a function of magnetic field applied along the c-axis (a) and in the ab plane (b) measured at different temperatures for the underdoped Bi2201 sample with p=0.12.

peratures, $T = 0.04 - 0.2$ K, in very high fields. This deviation cannot be related to the proximity of the superconducting transition since the behavior of $\rho_{ab}(T)$ in magnetic fields of 20 T and 27.5 T in Fig. 2(a) and (b) is identical. Moreover, the data at 27.5 T in Fig. 2(b) actually lie below the 20 T data. We interpret the observed onset of the saturation of ρ_{ab}, as a suppression of the localization by the magnetic field.

One can see in Fig. 2(a) that in the most underdoped sample with $p = 0.12$ at zero magnetic field there is the weak upturn in the region $3 - 4$ K, which we believe is a consequence of a competition between superconductivity and localization. To illustrate this, we show in Fig. 3 the magnetic-field dependence of R_{ab} for the same sample with $p = 0.12$ for temperatures from 40 mK to nearly 6 K for magnetic fields perpendicular (a) and parallel (b) to the *ab*-plane. The considerable difference in the $R_{ab}(H)$ curves between the two field orientations is a direct consequence of the anisotropy of the upper critical field in Bi2201 due to a difference in the orbital effect of the magnetic field on the one hand, and because the effect of the magnetic field on the localization is weaker for the parallel geometry on the other hand[50]. As can be seen from Fig. 3(b), at $T = 3$ K and 2.1 K, a negative magnetoresistance appears which results from the gradual suppression of localization effects by the magnetic field. This negative magnetoresistance also exerts some influence on the other $R_{ab}(H)$ curves at lower T, that is to say, the localization effects still persist. In the perpendicular geometry, the magnetic field suppresses rapidly the superconductivity and the competition between superconductivity and localization is not observed. Although the localization also exerts some influence on the $R_{ab}(H)$ curves in Fig. 3(a) (curves have a pronounced break-points in the derivative). Hence, we believe that the weak upturn in the zero-magnetic field ρ_{ab} in Fig. 2(a) is due to a competition between superconductivity and localization. Nevertheless, a sample inhomogeneity on atomic scale because of the heavy doping and proximity of the isolating phase cannot be ruled out as a possibility at this composition.

The negative magnetoresistance for the longitudinal geometry itself presents an additional difficulty for standard interaction theory. The same anomalous negative magnetoresistance for the longitudinal geometry at low temperatures has been observed previously in the non-superconducting Bi2201 single crystals by Jing *et al.*[49]. Since the authors[49] considered this phenomenon in detail, we will not discuss this topic further. However, it is important to note that in the second most underdoped sample with $p = 0.13$ the negative longitudinal magnetoresistance is not observed in spite of the fact that the ρ_{ab} at $T < 10$ K in Fig. 2(b) increases as $\log(1/T)$ and the localization persists. The data in Fig. 2 and Fig. 3 shows that the role of disorder in the field-induced normal

state of underdoped cuprates remains an open question. Further experiments are needed to reliably determine the low-temperature variation.

In contrast, $\rho_{ab}(T)$ for the slightly underdoped and overdoped samples with $p = 0.16$ Fig. 2(c) and 0.2 Fig. 2 (d) is constant below 5 K and clearly shows a metallic behavior in the normal state. This data is in full agreement with the behavior of $\rho_{ab}(T)$ in BSLCO and LSCO systems. Thus, it seems likely that the metal-insulator transition in Bi2201 lies in the underdoped region ($p < 0.16$) as for BSLCO. The observed metallic behavior gradually changes to an insulating behavior with decreasing carrier concentration.

In a 2D system the disorder parameter given by $k_F l$, where k_F is the Fermi wave vector and l the elastic scattering length, may serve as a measure of the disorder in the material[51]. From the residual resistivity $\rho_{ab}(T \to 0)$ in Fig. 2 and the lattice parameter c we determined the disorder parameter in the ab-plane $(k_F l)_{ab} \simeq 0.6, 7, 20,$ and 49 for samples with $p = 0.12$, 0.13, 0.16, and 0.2, respectively. For the samples with $p \geq 0.16$, $(k_F l)_{ab} >> 1$ and a true metallic conduction in the CuO_2 layers takes place, whereas the sample with p=0.12 clearly shows $\log(1/T)$ behavior starting from $T \simeq 30$ K where the value of ρ_{ab} is consistent with $(k_F l)_{ab} = 1.3$ (it is important to note that the Mott limit corresponds to $k_F l = 1$).

According to the optical data obtained by Tsvetkov *et al.*[52] on our Bi2201 single crystals, the effective mass in the ab-plane is $m^* = 3m_o$ where m_o is the free-electron mass. Using this value of m^* together with the carrier density we can calculate k_F and hence $l = 60$ and 145 $\AA$ at 10 K for the samples with $p = 0.17$ and 0.2, respectively. This clearly indicates that the optimally doped and underdoped Bi2201 crystals are clean superconductors. For these calculations we have assumed a cylindrically shaped Fermi surface with a highly anisotropic dispersion relation[53].

The large increase of ρ_{ab} is striking when compared with the small change in T_c when the hole doping p is changed from 0.13 to 0.12. This phenomena is not observed in the BSLCO system, which supports the suggestion of Ono *et al.*[37], that the disorder associated with (Sr,Bi) substitution is more harmful to the electronic system than the disorder due to (Sr,La) substitution. It is also possible that this results from the proximity of the isolating phase near the bottom of the phase diagram.

B. Out-of-plane resistivity ρ_c

Fig. 4 (main panel) shows the temperature dependence of the out-of-plane resistivity ρ_c at zero magnetic field for four single crystals shown in Fig. 1. The inset in Fig. 4 plots $\rho_c(T)$ on a semi-logarithmic scale to equally show the behavior of all the samples. As for the case of $\rho_{ab}(T)$, with decreasing p, the overall magnitude of ρ_c, increases as its "semiconducting" temperature dependence becomes less marked. The exception is the overdoped

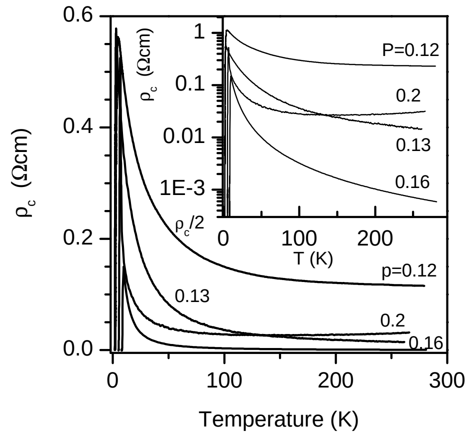

FIG. 4: c-axis resistivity ρ_c as a function of temperature for Bi2201 samples with different hole concentrations. The inset shows the same data plotted on a semi-log scale.

sample with $p = 0.2$, for which the ρ_c value is larger than ρ_c of the sample with $p = 0.16$ and this sample already shows a "metallic" temperature dependence of ρ_c at high temperatures. Such behavior at high temperatures is often observed in overdoped cuprates. The larger value of ρ_c in the overdoped sample Bi2201 is likely due to an excess of Bi and suggests a larger disorder in the electronic system compared to the in plane disorder in the same sample probed by ρ_{ab}. In all the underdoped crystals studied, we found that $\rho_c(T)$ at $H = 0$ T varies as a power law $T^{-\alpha}$ over the temperature range $T = 3-300$ K with $\alpha = 0.7 - 1.6$.

A $\log T$ plot of ρ_c at various fixed magnetic fields for samples from Fig. 4 is shown in Fig. 5 in order to emphasize again the low-temperature behavior. A strong magnetic-field induced suppression of the low-temperature upturn can be observed. In addition, $\rho_c(T)$ for the case of the slightly underdoped or overdoped crystals shows a tendency to saturate. One can see that the $\log(1/T)$ behavior of the ρ_c in the normal state gradually changes to a metallic-like behavior with increasing carrier concentration. The onset of this behavior in $\rho_c(T)$ moves to higher temperatures with increasing carrier concentration. Our data in Fig. 5 are in striking contrast to the behavior of $\rho_c(T)$ reported for the underdoped LSCO samples[54] and the slightly overdoped BSLCO single crystals[5], which exhibited a $\log(1/T)$ divergence in the normal state at $T \ll T_c$ (for temperatures up to 0.66 K). The metallic-like temperature dependence of the in-plane resistivity ρ_{ab} and a semiconducting-like behavior for the out-of-plane resistivity of ρ_c reported by Ando *et al.*[5] suggested that the c-axis transport is uncorrelated with the in-plane transport. On the other hand, the same $\log(1/T)$ divergence of $\rho_c(T)$ and $\rho_{ab}(T)$ in the underdoped LSCO samples gave the authors of Ref.[54] additional evidence against 2D localization. However, as is clear from Fig. 5, we do not have any evidence for a $\log(1/T)$ divergence at low temperatures in underdoped

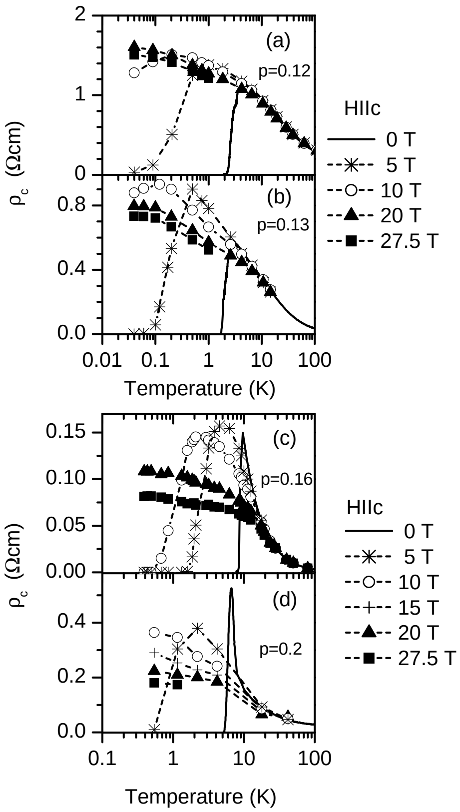

FIG. 5: ρ_c as a function of temperature and measured at various magnetic fields applied parallel to the c-axis for Bi2201 samples with different hole concentrations

Bi2201 single crystals, and the out-of-plane resistivity ρ_c of the slightly underdoped and overdoped Bi2201 single crystals below T_c in the highest applied fields shows almost no temperature dependence. This implies that the carrier-transport mechanism in the low-temperature limit, $T/T_c \to 0$, is the same for the ab and c directions. We note that Morozov *et al.*[1] have also observed a near saturation of ρ_c for a Bi2212 crystal in the temperature region $22.5 - 30$ K at 55 T.

A parameter more often used than ρ_c to characterize the interlayer coupling is the anisotropy of the resistivity ρ_c/ρ_{ab}. The largest anisotropy ratio found here is ρ_c/ρ_{ab} is 2.2×10^4 just above T_c. We find that the anisotropy ratio in zero-magnetic field for samples is strongly temperature dependent except for the most underdoped sample with $p = 0.12$ for which ρ_c/ρ_{ab} is significantly less and depends only slightly on temperature, probably due to the localization or enhanced disorder at this doping level. Such behavior is in agreement for the most part with the results of Wang *et al.*[55] and Ando *et al.*[5,37] previously reported for BSLCO samples and implies that at high temperatures the mechanisms governing transport along and perpendicular to the CuO_2 plane are different. However, the normal-state anisotropy ratio ρ_c/ρ_{ab} at low temperatures in very high magnetic fields becomes practically temperature independent for all samples. This behavior is in distinct contrast to Ref.[5] where ρ_c/ρ_{ab} of BSLCO crystals continued to increase below T_c providing evidence for the non-Fermi-liquid nature of the system. On the other hand, this result is consistent with data for the underdoped LSCO samples reported in Refs. [16,54]. The saturation of the ratio ρ_c/ρ_{ab} suggests that at low temperatures ρ_{ab} and ρ_c in very high magnetic fields are related, which is probably indicative of the anisotropic three-dimensional charge transport in this region induced by the magnetic field. In view of the remarkable difference between the temperature dependence of ρ_c/ρ_{ab} in Bi2201, BSLCO and LSCO, we do not want discuss here this topic more fully.

C. Pseudogap

According to Ref.[1], the interlayer transport results from a tunneling process and quasiparticle tunneling dominates at higher fields. Since ρ_c can give information about the quasiparticle density of states in the presence of a pseudogap, below we will discuss the ρ_c magnetoresistivity at high fields in our samples. The suppression of a semiconducting-like temperature dependence for $\rho_c(T)$ can be interpreted as the magnetic-field induced suppression of the pseudogap, previously observed at temperatures above 5 K for slightly underdoped Bi2201 crystals with $T_c = 9.5$ K[31] and in highly overdoped Bi2212 single crystals[56] at $T > 20$ K ($T_c \approx 60$ K).

In Fig. 6 we plot $\rho_c(H)$ versus magnetic field for four Bi2201 single crystals. For completeness in Fig. 6 (c), we also display our data for the slightly underdoped ($p = 0.16$) sample[31]. The inset in Fig. 6 (c) shows the relative variation $\Delta\rho_c/\rho_{c0} = [\rho_c(H,T) - \rho_c(0,T)]/\rho_c(0,T)$ at different temperatures for both configurations at a magnetic field $H = 28$ T. After the magnetic field induced onset suppression of superconductivity all samples show a positive magnetoresistance at low fields. The maximum in $\rho_c(H)$ observed at higher fields is followed by a region of negative magnetoresistance. Fig. 6 clearly shows the difference between the behavior of $\rho_c(H)$ in the underdoped and overdoped crystals. At low temperatures ρ_c in the overdoped regime shows a much stronger negative magnetoresistance compared to that observed in the underdoped regime. Such a behavior of $\rho_c(H)$ has already been intimated in the Bi2212 system[32]. However, such a large difference between the underdoped and overdoped regimes in the slope of the negative magnetoresistance in Fig. 6, has not previously been observed. Furthermore, in the heavily underdoped sample ($p = 0.12$) after an increase of ρ_c at low fields due to the gradual suppression of superconductivity, ρ_c decreases almost linearly with increasing magnetic field up to $\simeq 28$ T even at very low temperatures in contrast to the power-law field dependence previously reported in references [1,32].

In Ref. [32] it was found that the field at which the excess "semiconducting" resistivity $\Delta\rho_c(T)$ vanishes corresponds to the pseudogap closing field H_{pg}. A fit

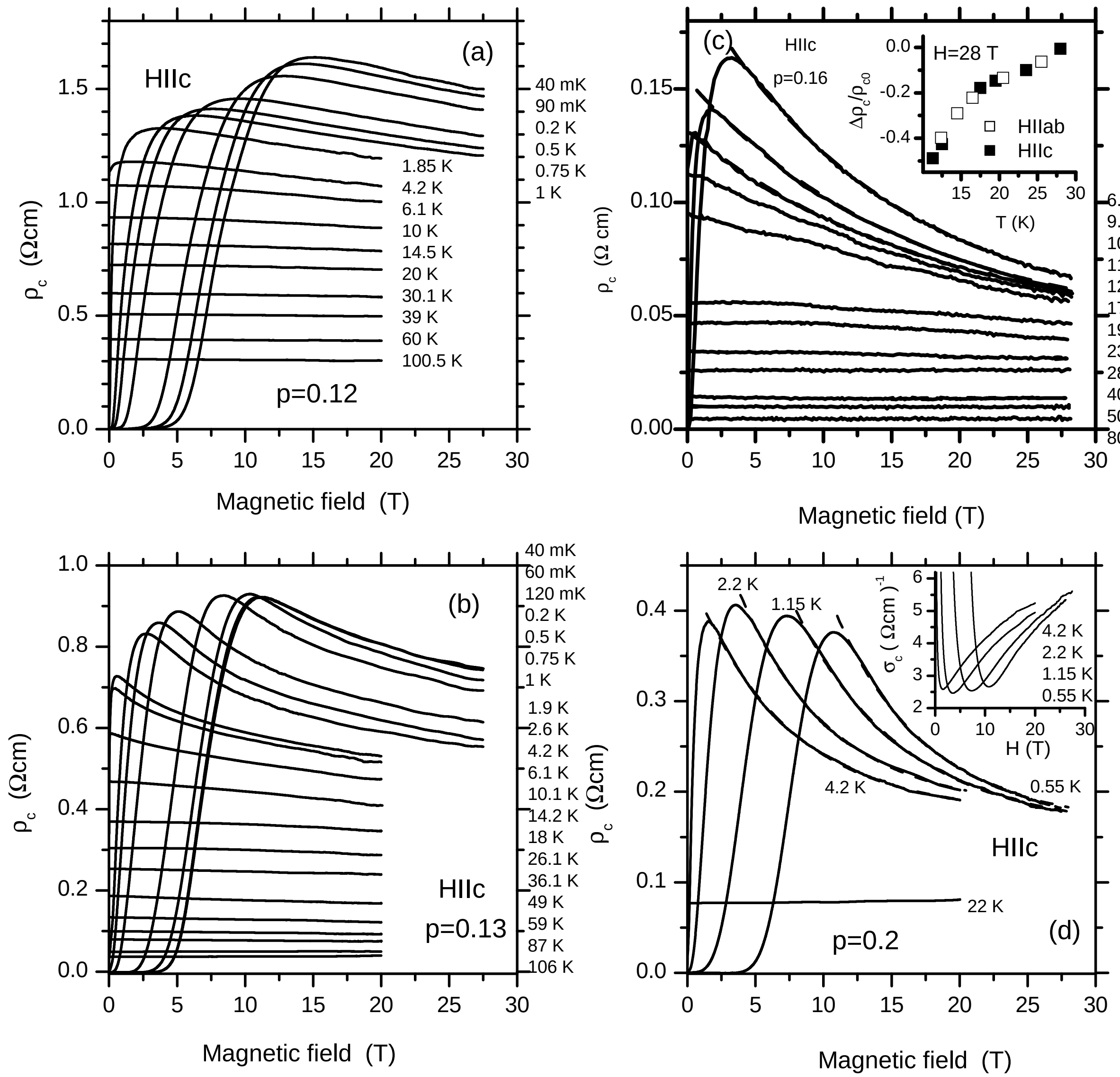

FIG. 6: ρ_c as a function of magnetic field applied parallel to the c-axis for different temperatures for Bi2201 samples with various hole concentrations. The inset in (c) shows the relative variation $\Delta\rho_c/\rho_{c0} = [\rho_c(H,T) - \rho_c(0,T)]/\rho_c(0,T)$ at different temperatures for both configurations at a magnetic field $H = 28$ T. The inset in (d) shows the c-axis conductivity for the same sample ($p = 0.2$).

to the power-law dependence of $\Delta\rho_c(H)$ for magnetic fields above the maximum in $\rho_c(H)$ at different temperatures allowed the authors of Ref.[32] to find the field at which $\Delta\rho_c$ vanishes and evaluate $H_{pg}(T)$ beyond the available 60 T. Based on this suggestion, we tried to fit to a near linear field dependencies of $\Delta\rho_c(H)$ in a log-log plot for $p = 0.12$ and $p = 0.13$ in order to evaluate H_{pg} at low temperatures in underdoped samples. This evaluation gives exaggeratedly large values for $H_{pg} \approx 2000 - 3000$ T. In Ref.[32] it has also been found that H_{pg} and T^* are related through the Zeeman-like expression $g\mu_B H_{pg} = k_B T^*$, where $g = 2$ is the electronic g-factor, μ_B the Bohr magneton, and k_B the Boltzmann constant. In our case such an analysis leads to physically meaningless values $T^* = 2700-4000$ K. Other extrapolation polynomial fits gave the same physically meaningless values of H_{pg}. These results probably indicate that the method suggested in Ref. [32] for evaluating H_{pg} is unsuccessful in case of underdoped Bi2201 samples.

We have tried to use such an extrapolation fit to our $\rho_c(H)$ data for overdoped Bi2201. Fig. 7 shows a log-log plot of $\rho_c(H)$ at various fixed temperatures for the overdoped sample with $p = 0.2$. It can be seen that the dashed straight lines, which are extensions of the linear dependencies, point to the limiting value[32] of H_{pg}, corresponding to the intersection at 25 T. If $H_{pg} \simeq 25$ T, then using the Zeeman-like expression T^* is found to be $\simeq 34$ K. In the overdoped Bi2212 samples, the negative

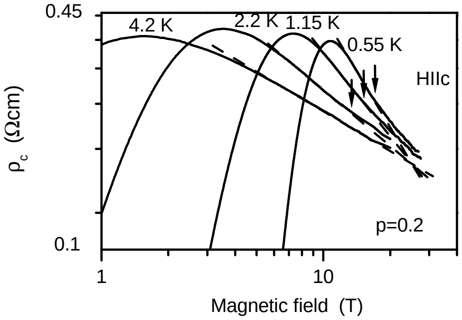

FIG. 7: Log-log plot of ρ_c versus magnetic field for various temperatures for the overdoped Bi2201 samples.

magnetoresistance disappears at the same temperature at which the zero-field $\rho_c(T)$ deviates from its characteristic linear (metallic) high-temperature dependence[32,56]. This temperature in Ref. [57] was identified as the pseudogap closing (opening) temperature T^*. However, as can be seen from Fig.4, the zero-field $\rho_c(T)$ of sample with $p = 0.2$ deviates from a metallic linear (high) temperature dependence at $T \simeq 140$ K, so that H_{pg} should be $\simeq 100$ T. The sample with $p = 0.16$ does not show any linear T dependence (metallic state) up to 270 K (suggesting that H_{pg} should be > 200 T). This result is clearly inconsistent. Moreover, in Fig. 7 one can see that even in case of overdoped Bi2201 samples there is only a finite range of magnetic field for which the $\rho_c(H)$ data can be described by a power-law[32,56] dependence H^α (dashed lines). This result indicates that the method suggested in Ref. [32] for evaluating H_{pg} is unsuccessful in case of overdoped Bi2201 samples also.

Once the magnetic field at which the negative magnetoresistance vanishes is identified with the pseudogap closing field H_{pg}, our results clearly show that in the Bi2201 samples investigated here, the pseudogap closing temperature T^* does not agree with the temperature at which the zero-field semiconducting-like temperature dependence of ρ_c changes into a metallic dependence at higher temperatures as in the overdoped Bi2212[32]. Since the metallic-like linear temperature dependence of the ρ_c at $H = 0$ T is a consequence of the high doping of the samples which is inevitably accompanied by a severe degradation of samples quality, we are unable to reach an unambiguous conclusion concerning the relation of H_{pg} with the deviation from the linear temperature dependence of ρ_c.

However, on the other hand, in our slightly underdoped ($p = 0.16$) and overdoped ($p = 0.2$) Bi2201 crystals the negative magnetoresistance vanishes and the magnetoresistance changes sign at ≈ 28 K, the inset in Fig. 6(c), and at ≈ 22 K, Fig.6(d). Thus, T^* should be close to these temperatures. According to the the Zeeman-like expression (Ref.[32,56]) the pseudogap closing field scales with T^* as $g\mu_B H_{pg} = k_B T^*$ which implies that H_{pg} should be $\simeq 21$ T and $\simeq 16$ T, respectively.

In the slightly underdoped ($p = 0.16$) and overdoped ($p = 0.2$) samples, the strong negative magnetoresistance rapidly weakens [Fig. 6(c) and (d)] and clearly shows a saturation at high fields after more than a two-fold decrease. When the temperature-dependent data in Fig. 5(c) and (d) are compared with Figs. 6(c) and (d), it can be concluded that the observed negative magnetoresistance corresponds to a suppression of the semiconducting-like behavior in $\rho_c(T)$, which can in turn be interpreted as the magnetic-field induced suppression of the pseudogap. In previous measurements[33] we have shown that all $\rho_c(H)$ curves for Bi2201 single crystal with $T_c \simeq 7$ K (overdoped) have a pronounced break-point in the derivative well above the $\rho_c(H)$ peak, which shifts to higher fields with decreasing temperature and at $T \simeq T_c$ disappears. The field position of these break-points in the derivative coincide with the H^*_{c2} values determined from the $\rho_{ab}(H)$ curves. The values of H at which the log-log plot of $\rho_c(H)$ deviates from a linear magnetic field dependence in Fig. 7 (shown by arrows) are in close agreement with the H^*_{c2} values for a Bi2201 sample[33] with $T_c \simeq 7$ K. As has been shown in Refs.[32,56], H_{pg} in Bi2212 does not depend on temperature for $T < T_c$ and as $T \to 0$ H_{pg} and the upper critical field, H_{c2}, coincide. This suggests again that the intersection points of the dashed straight lines in Fig. 7 is not H_{pg} as observed in Bi2212. On the other hand, if H_{pg} is determined from the disappearance of the negative magnetoresistance and, as pointed out above, $H_{pg} \approx 21$ T ($p = 0.16$) and $H_{pg} \approx 16$ T ($p = 0.2$), so that H_{pg} and H^*_{c2} in Bi2201 are closely linked as in Bi2212.

Yurgens *et al.*[58] measured the intrinsic-tunneling spectra of a La-doped Bi2201 (T_c=32 K) at $T = 4.5$ - 300 K in order to determine the pseudogap phase diagram. Their phase diagrams show that for samples with p=0.16 and under, the pseudogap closing temperature T^* is over 300 K. These temperatures lie outside the range of our measurements. While for the overdoped sample with p=0.2, the value of T^*=22 K found in our work agrees well with the pseudogap phase diagram of Yurgens *et al.*[58].

The negative magnetoresistance observed in our experiments show a characteristic exponential decrease with magnetic field. Fig. 6(c) and (d), show numerical fits (the dashed curves) calculated using the functional form $\rho_c(H,T) = \rho_{c0} + a\exp(-H/bT)$, where a and b are constants. Our data in the slightly underdoped, optimally doped and overdoped regimes are well described by such a functional form. The possibility to describe $\rho_c(H)$ by an exponential expression in H/T implies the magnetic-field couples to the pseudogap via the Zeeman energy of the spin degrees of freedom[31,32].

In previous measurements of near optimally doped Bi2201 single crystals[31], we have found an isotropic behavior of the normal-state magnetoresistance with respect to the orientation of the magnetic field (perpendicular and parallel to the CuO_2 planes) which showed that only the effect of the magnetic field on the spins (Zeeman effect) is important in the normal state. Here a negative magnetoresistance is observed for both geome-

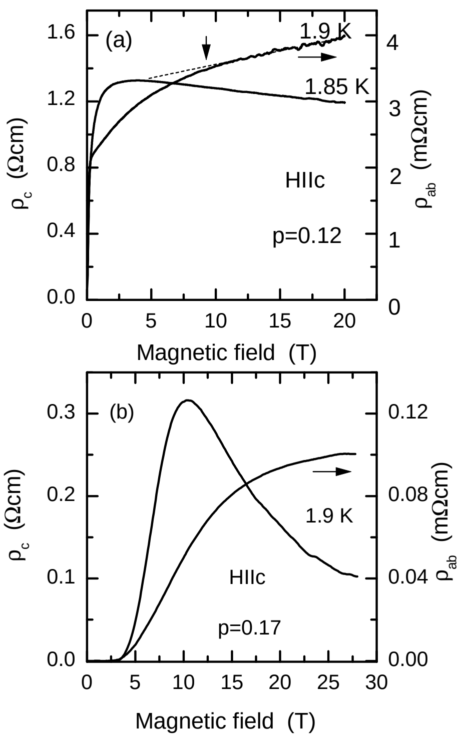

FIG. 8: ρ_c and ρ_{ab} measured as a function of magnetic field applied along the c-axis around T=1.9K for the (a) underdoped and (b) optimally doped Bi2201 samples.

tries ($\mathbf{H} \parallel \mathbf{c} \parallel \mathbf{J}$ and $\mathbf{H} \perp \mathbf{c} \parallel \mathbf{J}$) in all investigated crystals. In contrast to the magnetoresistance in the superconducting state, the normal-state magnetoresistance of ρ_c is independent of the field orientation with respect to the current direction. For the slightly underdoped sample ($p = 0.16$), this behavior can be seen in the inset in Fig. 6 (c).

We observed the same behavior for the heavily underdoped sample ($p = 0.12$). The similarity in the normal-state data for the two field orientations, probably excludes an explanation of the normal state negative out-of-plane magnetoresistance in terms of superconductivity.

It should especially be pointed out that in slightly underdoped, optimally doped and overdoped samples after the field induced suppression of the superconductivity and pseudogap for ρ_c in high fields, the value of ρ_c (after the saturation of the magnetoresistance) remains much higher than the expected un-gapped value. *A semiconducting-like temperature dependence of the out-of-plane resistivity* ρ_c *is partly conserved even after the suppression of the negative magnetoresistance at* $H > H_{pg}$. It seems reasonable to conclude that the semiconducting-like temperature dependence of ρ_c is controlled not only by the magnetic-field-sensitive pseudogap.

In previous measurements[33] we have pointed out that in overdoped samples ($T_c = 7$ K) the maxima of $\rho_c(H)$ coincide with the field positions of $\rho_{ab}(H) = 0.4\rho_{ab}^n$ where ρ_{ab}^n is the normal-state resistivity. The observed maximum in $\rho_c(H)$ is a property of the mixed state and results from a competition between the "semiconducting" behavior of ρ_c and the superconducting transition. In Fig. 8 we display the resistive transitions of the heavily underdoped [(a), p=0.12] and optimally doped [(b), p=0.17] samples in a magnetic field $\mathbf{H}\|\mathbf{c}\|\mathbf{J}$ at temperatures ≈ 1.9 K. It can be see that after the maximum in $\rho_c(H)$, the $\rho_{ab}(H)$ curves still show a strong positive magnetoresistance, clearly originating from the superconducting state. In this range of magnetic fields a superconducting gap still persists and part of the current along the c-axis is a quasiparticle tunneling current. In the underdoped sample with lower T_c the superconducting gap is small and closes rapidly when magnetic field is applied. The resistive transition to the normal state is completed at ≈ 10 T (the weak increase of the normal-state ρ_{ab} resistivity is due to a magnetoresistance contribution at high magnetic fields). The negative magnetoresistance at $H > 10$ T displayed by $\rho_c(H)$ in Fig. 8(a) is isotropic and is due to the magnetic-field induced suppression of the pseudogap. Since in the underdoped samples the magnitude of the pseudogap is large, the effect of the magnetic field is small and the negative magnetoresistance does not saturate in the available magnetic field range [Fig. 6(a),(b)]. In the optimally doped [Fig. 8(b)], slightly underdoped and overdoped [Fig. 6(c), (d)] samples because H_{c2} is large, the major contribution to the anisotropic negative magnetoresistance in the $\rho_c(H)$ curves is due to the gradual decrease of the superconducting gap (in Fig. 7, the end of the superconducting transition is indicated by arrows). Since in these samples the magnitude of the pseudogap is small, the negative magnetoresistance connected with the pseudogap rapidly saturates following the superconducting transition [Fig. 6 (c), (d)]. However, as previously pointed out the value of ρ_c remains much higher than the expected un-gapped value. Recently it has been shown that the negative magnetoresistance in the superconducting state can also be described by the Zeeman-like expression[59]. This explains why it is possible to describe completely all curves $\rho_c(H)$ in Fig. 6(c), (d) using an expression which is exponential in H/T in both the superconducting and normal states.

IV. CONCLUSION

We have presented the temperature dependence for both in-plane ρ_{ab} and out-of-plane ρ_c resistivities and magnetoresistivities $\rho_{ab}(H)$ and $\rho_c(H)$ in hole-doped La-free Bi2201 cuprate for a wide doping range and over a wide range of temperatures down to 40 mK. We have shown that the temperature and magnetic field dependence of the in-plane and out-of-plane resistivities are determined by the localization, the superconducting gap and the normal-state pseudogap. The data suggest that the metal-to-insulator crossover in Bi2201 lies in the un-

derdoped region ($p < 0.16$). The metallic behavior of $\rho_{ab}(T)$ gradually changes to insulating behavior with decreasing carrier concentration. We did not observe any evidence for a $\log(1/T)$ divergence of ρ_{ab} and ρ_c at very low temperatures in underdoped Bi2201 single crystals. The out-of-plane resistivity ρ_c of the slightly underdoped and overdoped samples below T_c in the highest applied fields shows almost no temperature dependence. Our data strongly suggest that the negative out-of-plane magnetoresistance appears to be governed by different mechanisms; the main contribution comes from the transition to the normal state which gives rise to a strong magnetic field dependence, while the non-superconducting pseudogap, shows a much weaker magnetic field dependence and therefore only gives a small contribution to the negative out-of-plane magnetoresistance. A semiconducting-like temperature dependence of the out-of-plane resistivity ρ_c is conserved in part even after the suppression of the negative magnetoresistance and at magnetic fields above the pseudogap closing field H_{pg}. Our data support that the pseudogap does not correlate with the existence of superconducting gap.

Acknowledgments

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